

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of:

Kiyoshige MURAOKA

Group Art Unit: 1791

Serial Number: 10/679,406

Examiner: Geoffrey L. Knable

Filed: October 29, 2003

For: TUBELESS TIRE

DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents  
P.O. Box 1450  
Alexandria , Virginia 22313-1450

Sir:

Kiyoshige Muraoka residing at 4-3-2, Oyashikidori,  
Nagata-ku, Kobe-shi, Hyogo, Japan duly deposes and says:

1. That he graduated from Department of Applied  
Chemistry, School of Engineering, University of Hyogo, Hyogo, Japan,  
in the year 1983, and he received the degree of Master of Process  
Engineering from Graduate School of Osaka University, Osaka, Japan  
in the year 1985;

2. That since 1985, he has been employed in the capacity  
of Sumitomo rubber Industries, Ltd.;

3. That from 1985 he has been engaged in development of  
rubber compositions for tires.;

4. That he has read and is familiar with the instant  
application for United States Letters Patent and Office Action thereto  
mailed November 29, 2007.; and

5. That he has made experiments in order to show that a tubeless tire having a gas barrier layer and an inner liner layer comprising a rubber composition containing an inorganic filler defined in the present invention described in USSN 10/679,406 is excellent in rate of decrease in pressure after machining and appearance of the gas barrier layer, and that a tubeless tire having a gas barrier layer and a carcass layer comprising a rubber composition containing an inorganic filler defined in the present invention described in USSN 10/679,406 is excellent in tire durability, rate of decrease in pressure after machining and appearance of the gas barrier layer.

### **Experiments**

<Process for preparing coating solution for gas barrier layer>

Coating solution 1

Synthetic mica (tetrasilic mica (Na-Ts); available from Topy Industries, Limited) was dispersed in ion exchanged water so as to become 0.65 % by weight to obtain an inorganic layered compound dispersion solution (solution A). The average particle size of the synthetic mica (Na-TS) was 977 nm and the average aspect ratio was 1043. Polyvinyl alcohol (PVA210; available from Kuraray Co., Ltd., weight percentage of hydrogen bond groups or ionic groups per weight unit of resin: 38 %, hydrolyzation degree: 88.5 %, polymerization degree: 1000) was dissolved in ion exchanged water so as to become 0.325 % by weight to obtain a resin solution (solution B). Solution A and solution B were mixed so that the respective solid content ratio (volume ratio) was inorganic layered compound/resin = 3/7 to obtain coating solution 1 for the gas barrier layer.

<Preparation of rubber composition>

Each sample rubber composition (rubber compositions J to O, J' to O', U, W, U' and W') was prepared with the materials and processing methods described below.

(Materials)

NR: RSS #3 available from Tech Bee Hang Co., Ltd.

Br-IIR: Exxon Bromobutyl 2255

Sodium-bentonite: Kunipia F (clay-type mineral with a primary particle size of 100 to 2000 nm, average aspect ratio of 320 and swellability of at least 45 ml/2 g) available from Kunimine Industries

GPF: Seast V available from Tokai Carbon Co., Ltd.

HAF: Diablack H available from Mitsubishi Chemical Corporation

Silica: Ultrasil VN3 ( $N_2SA$ : 210 m<sup>2</sup>/g) available from Degussa Co.

Silane coupling agent: Si69 (bis(3-triethoxysilylpropyl)tetrasulfide) available from Degussa Co.

Resin: ESCOREZ 1102 available from Esso

Oil A: JOMO Process X-140 available from Japan Energy Corporation

Oil B: Machine oil 22 available from Showa Shell Sekiyu K.K.

Stearic Acid: Stearic acid available from NOF Corporation

Zinc oxide: Zinc Oxide Type 1 available from Mitsui Mining and Smelting Co., Ltd

Sulfur: powdery sulfur available from Tsurumi Chemicals Co., Ltd.

Vulcanization Accelerator NS: Nocceler NS  
(N-tert-butyl-2-benzothiazylsufenamide), available from Ouchi Shinko Chemical Industrial Co., Ltd.

Vulcanization Accelerator DM: Nocceler DM available from Ouchi

Shinko Chemical Industrial Co., Ltd.

Vulcanization Accelerator D: Nocceler D (N,N'-diphenyl guanidine),  
available from Ouchi Shinko Chemical Industrial Co., Ltd.

(Processing method)

According to the composition formula shown in Tables A and B, the materials other than sulfur, zinc oxide and the vulcanization accelerator were kneaded in a BR-type Banbury to prepare a master batch and then the master batch and sulfur, zinc oxide and the vulcanization accelerator were kneaded with an 8 inch roll to obtain each sample rubber composition (rubber composition J to O, J' to O', U, W, U' and W'). The obtained rubber composition was press vulcanized at 170°C for 15 minutes to obtain a vulcanized object.



TABLE B

Rubber Composition	U	W	U'	W'
Composition (parts by weight)				
Natural Rubber	40	40	40	40
Br-IIR	60	60	60	60
HAF	20	-	20	-
Sodium-bentonite	5	5	5	5
Oil A	10	10	10	10
Silica	20	50	-	-
Silane Coupling Agent	2	5	2	5
Resin	2	2	2	2
Oil B	-	-	-	-
Stearic Acid	2	2	2	2
Zinc Oxide	3	3	3	3
Sulfur	1.5	1.5	1.5	1.5
Vulcanization Accelerator NS	1	1	1	1
Vulcanization Accelerator D	1	1	1	1

### Experimental Examples 1 to 12

#### <Evaluation as rubber composition>

After coating solutions 1 was formed into a film by casting on the various rubber samples obtained by rubber compositions J to O and J' to O', according to the compounding formula shown in Table A, thermal treatment was conducted for 10 minutes at 100°C. The film thickness obtained by the treatment was approximately 5  $\mu\text{m}$ . The air permeability evaluation was conducted with respect to the rubber. The results are shown in Table C.

In each Experimental Example, "1" indicates that coating solution 1 for the gas barrier layer was formed into a film in the gas barrier composition of Table A.

#### (Air permeability evaluation)

The air permeability coefficient was measured according to JIS K7126 "Testing method of gas permeability of plastic films and sheets (method A)" with air as the test gas (nitrogen:oxygen = 8:2) and 25°C as the test temperature. The air permeability is decreased the lower the number value is.

The air permeability coefficient is preferably at most 30 ( $\times 10^{-11} \text{cc}\cdot\text{cm}/\text{cm}^2\cdot\text{sec}\cdot\text{cmHg}$ ). More preferably, the air permeability coefficient is in the range of at most 20. When the air permeability coefficient is more than 30, low permeability may not to be obtained.

#### <Evaluation as tire>

A 195/65R14 tire was prepared using the above rubber

composition as the inner liner layer and then vulcanized.

The coating solution for the gas barrier layer was applied to the inner face of the inner liner layer of the vulcanized tire using a certain spray gun and then dried. Then, thermal treatment was conducted for 10 minutes at 100°C. The film thickness obtained by the treatment was approximately 2 µm. The evaluation of the rate of decrease in tire pressure and the following pressure test and evaluation of appearance after testing with a tire machine were conducted with respect to the tire. The results are shown in Table C.

(Evaluation of rate of decrease in tire pressure)

The above tire with an initial pressure of 100 kPa was left for 3 months in room temperature of 25°C under a no-load condition and the pressure was measured every 4 days. When  $t$  is the number of days passed,  $P_0$  is the initial pressure and  $P_t$  is the pressure measured when  $t$  days have passed, the value of  $\alpha$  is found by using function:

$$P_t/P_0 = \exp(-\alpha t).$$

The obtained  $\alpha$  and  $t = 30$  were substituted into the following equation and  $\beta$  was obtained. The value of  $\beta$  was assumed to be the rate of decrease in pressure per month (%/month).

$$\beta = \{1 - \exp(-\alpha t)\} \times 100$$

The rate of decrease in pressure of the tire is preferably at most 2.5 (%/month). When more than 2.5 %, decrease in tire pressure becomes noticeable.

(Pressure test and evaluation of appearance after testing with tire



machine)

An indoor drum test was conducted using a specific tire under conditions of running speed of 80 km/hour, pressure of 190 kPa and load of 646 kg and the tire was run for 30,000 km. The appearance of the inner liner layer was checked for tears and wrinkles in the gas barrier layer thin film. Then, with respect to the tire, the test for the rate of decrease in tire pressure was conducted again.

TABLE C

Experimental Example													
	1	2	3	4	5	6	7	8	9	10	11	12	
ber Composition	K	M	O	J	L	N	K'	M'	O'	J'	L'	N'	
Barrier Layer Composition	1	1	1	1	1	1	1	1	1	1	1	1	
Permeability Coefficient O <sub>2</sub> ·cc·cm/cm <sup>2</sup> ·sec·cmHg)	5	5	3	8	7	4	5	5	3	8	7	4	
% of Decrease in Pressure (month)	1.5	1.5	1.3	1.9	1.9	1.6	1.5	1.5	1.3	1.9	1.9	1.6	
% of Decrease in Tire Pressure or Machining (%/month)	1.8	1.5	1.4	2.1	2.0	1.7	2.0	2.0	2.0	2.4	2.3	2.1	
variance of Gas Barrier layer of Tire	No Wrinkles	No Wrinkles	No Wrinkles	Few Wrinkles	No Wrinkles	No Wrinkles	Few Wrinkles	Few Wrinkles	Few Wrinkles	Scattered Wrinkles	Few Wrinkles	Few Wrinkles	

### Experimental Examples 13 to 16

#### <Evaluation as rubber composition>

After coating solutions 1 was formed into a film by casting on the various rubber samples obtained by rubber compositions U, W, U' and W', according to the compounding formula shown in Table B, thermal treatment was conducted for 10 minutes at 100°C. The film thickness obtained by the treatment was approximately 5  $\mu\text{m}$ . The air permeability evaluation was conducted with respect to the rubber. The results are shown in Table D.

In each Experimental Example, "1" indicates that coating solution 1 for the gas barrier layer was formed into a film in the gas barrier composition of Table D.

#### <Evaluation as tire>

A 195/65R14 tire was prepared using the above rubber composition as the carcass layer and then vulcanized. In Experimental Examples 13 to 16, an inner liner layer was not formed.

The coating solution was applied to the inner face of the vulcanized tire using a certain spray gun and then dried. Then, thermal treatment was conducted for 10 minutes at 100°C. The film thickness obtained by the treatment was approximately 2  $\mu\text{m}$ . The evaluation of the rate of decrease in tire pressure, the pressure test and evaluation of appearance after testing with a tire machine and the following tire durability test were conducted with respect to the tire. The results are shown in Table D.

(Tire durability test)

An indoor drum test was conducted using a specific tire under conditions of running speed of 80 km/hour, pressure of 190 kPa and load of 464 kg. The running results are represented by the distance run from when running is begun until abnormality such as burst or change in the tire appearance (swelling) is observed. The durability is superior the longer the running distance is.

TABLE D

	Experimental Example			
	13	14	15	16
Rubber Composition	U	W	U'	W'
Gas Barrier Layer Composition	1	1	1	1
Air Permeability Coefficient ( $\times 10^{-11} \text{cc}\cdot\text{cm}/\text{cm}^2\cdot\text{sec}\cdot\text{cmHg}$ )	4	4	4	4
Inner Liner (Rubber Composition Z) (Air Permeability Coefficient : $19 \times 10^{-11} \text{cc}\cdot\text{cm}/\text{cm}^2\cdot\text{sec}\cdot\text{cmHg}$ )	None	None	None	None
Rate of Decrease in Pressure (%/month)	2.0	2.0	2.0	1.9
Tire Weight (kg)	8.61	8.61	8.60	8.60
Results of Durability Test (10,000 km)	3.0 No Breakage	3.0 No Breakage	3.0 No Breakage	1.5 Swelling of Sidewall area
Rate of Decrease in Tire Pressure after Machining	2.1	2.0	2.8	Unable to Measure
Appearance of Gas Barrier layer Inside Tire	No Wrinkles	No Wrinkles	Few Wrinkles	-

## **Result and Discussion**

As shown in Table C, in Experimental Examples 1 to 6 in which rubber compositions J to O containing silica as inorganic filler were used, excellent results were obtained regarding rate of decrease in pressure after machining and appearance of the gas barrier layer, in comparison to Experimental Examples 7 to 12, in which rubber compositions J' to O' containing no inorganic filler such as silica were used.

In addition, as shown in Table D, in Experimental Examples 13 and 14 in which rubber compositions U and W containing silica as inorganic filler were used, excellent results were obtained regarding tire durability, rate of decrease in pressure after machining and appearance of the gas barrier layer, in comparison to Experimental Examples 15 and 16 in which rubber compositions U' and W' containing no inorganic filler such as silica were used.

Consequently, it is proved that the tubeless tire having the gas barrier layer and the inner liner layer comprising the rubber composition containing the inorganic filler defined in the present invention described in USSN 10/679,406 is excellent in rate of decrease in pressure after machining and appearance of the gas barrier layer, and, in addition, the tubeless tire having and the gas barrier layer the carcass layer comprising the rubber composition containing the inorganic filler defined in the present invention described in USSN 10/679,406 is excellent in tire durability, rate of decrease in pressure after machining and appearance of the gas barrier layer.

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This 24th day of April, 2008

by Kiyoshige Muraoka  
Kiyoshige Muraoka

We, the undersigned witnesses, hereby acknowledge that Kiyoshige Muraoka is personally known to us and did execute the foregoing Declaration in our presence on:

Date: April 24, 2008

Witness Shigen Fujimara

Date: April 24, 2008

Witness Naoki Fujita